

Heat-related Changes of Density, P-wave Velocity, and Surface Hardness of Granite

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Abstract

Low- and intermediate-level radioactive waste is stored in the National Radioactive Waste Repository in Bataapáti in Hungary. The repository is located in the Carboniferous Mórággy Granite Formation. This paper focuses on heat-related changes of physical properties such as bulk density, P-wave velocity, P-wave modulus, and Duroskep surface hardness of the dominant lithology: monzogranite. Cylindrical specimens were tested at laboratory conditions (22 °C) and were heat-treated up to 250 °C, 500 °C, and 750 °C. The properties were measured before and after the thermal strain. After heat-treatment, the monzogranite samples became brownish, and at 750 °C, cracks appear at the surface of the specimens. Laboratory test results show that bulk density values slightly decrease from room temperature to 250 °C treatment and further dropped at 500 °C and especially at 750 °C. P-wave velocity values and the connected P-wave modulus tend to decrease from room temperature to 750 °C significantly. Duroskep rebound values show slight declines in the surface strength of the specimens until 500 °C, and then a drastic decline at 750 °C. Heat treatment tends to alter the physical properties of the monzogranite. From room temperature to 500 °C, a slight but apparent decrease between 500 °C and 750 °C significant reductions in the bulk density, P-wave velocity, and Duroskep values. Behind the physical alterations are the different thermal-induced expansion of minerals and mineral alteration at elevated temperatures.

Keywords

granite, thermal-treatment, bulk density, P-wave velocity, duroskep

1 Introduction

Granite is a commonly used material in construction and applied earth science studies [1–3]. It is preferred for general construction use as raw material in civil engineering practice and for building high-volume structures with special requirements. The increasing use of this rock type is the host rock of a subsurface radioactive waste disposal facility, implemented or planned in many countries [4, 5].

Low- and intermediate-level radioactive waste has been stored at Bataapáti National Radioactive Waste Repository Eastern-Mecsek (Hungary). The host rock is the Carboniferous Mórággy Granite Formation.

One of the most important safety considerations for deep geological radioactive waste storage is that excessive temperatures are avoided, and the facility works at normal temperatures. However, during certain failure events (such as heat caused by radioactive waste decay and sudden fire events), the temperature of the facility can reach several hundred degrees of Celsius [6–9]. The consequences of such damage events need to be investigated.

The thermomechanical properties of granitoid rocks have been receiving more attention in the last few decades. Physical and geochemical parameters of the rock are well-known, thermal-physical property changes are increasingly studied.

Heuze highlighted that strength, density, thermal expansion, latent heat, and melting temperature are well researched; however, topics of physical parameter changes above the 576 °C transition of quartz, especially above 700 °C, were less well studied [10]. Another study focused on the dynamic mechanical behavior of thermally treated granites such as dynamic strength and dynamic P-wave modulus from room temperature up to 800 °C [11]. Non-destructive and destructive mechanical testing of heat-treated (up to 600 °C) specimens of the Mórággy Granite Formation suggested that bulk density and ultrasonic pulse velocity are good indicators of heat-related physical changes of these granites [12]. The latter is significant since the temperature can cause irreversible mechanical

changes that can influence the stability of underground storage facilities [13, 14]; hence the stability of radioactive waste disposal sites is primarily controlled by mechanical parameters of the host rock [15].

This paper presents the heat-related changes of physical parameters of monzogranite specimens of the Mórággy Granite Formation. The observed values were mass-, volume-, bulk density, P-wave velocity, and Durosokop rebound changes from room temperature to 250 °C, 500 °C, and 750 °C of heat treatment.

2 Materials and methods

The host formation of the repository is a granitoid rock body mainly formed from monzogranite, monzonite, hybrid rocks, veins, and xenoliths [16]. The tested specimens belong to the porphyritic monzogranite group of the Mórággy Granite Formation (Fig. 1).

The porphyritic monzogranite is a dark reddish grey, phenocrystalline, medium to coarse-grained, inequigranular fresh rock formation with main minerals of quartz K-feldspar and biotite. The pink K-feldspar crystals are megacrysts with a maximum of 3 cm in diameter. The minerals are hypidiomorphic to rarely xenomorphic. The rock type has an undirected texture in the examined zone of the core drill. [16]

Altogether, 24 pieces of cylindrical samples with an average diameter of 4.73 cm and a 2:1 (height/width) ratio were prepared for treatment and laboratory tests. All in all 4 groups (22 °C, 250 °C, 500 °C, 750 °C) were examined, in each group, 6 specimens were tested to determine statistical values.

The samples were cut to shape from NQ-sized cores with a water-cooling diamond cutting disc (Fig. 2(a)). The samples were dried out to an air-dry state then mass, volume (i.e., bulk density), ultrasound velocity measurements, and surface hardness measurements were carried out (Fig. 3(a) and (b)). Bulk density values were measured according to EN 1936:2007 European Normative [17]. Ultrasound P-wave velocity values of the specimens were recorded with a PUNDIT device according to EN 14579:2005-01



Fig. 1 Macroscopic texture of porphyritic monzogranite samples of the Mórággy Granite Formation

standard [18]. The surface strength was measured with a non-standardized Durosokop test with a frame for testing cylindrical samples [19].

The heat-treatment of the samples was carried out in a Carbolite ABA 7/35 electric oven (Fig. 2(b)). During the treatment, heating was performed at 20 °C/min until the desired 250 or 500 or 750 °C was reached. The heat treatment was then carried out for 4 hours to allow the appropriate thermal homogenization to occur; after then, the samples were cooled to room temperature at an average rate of 5 °C/min. The samples were then re-tested for bulk density, ultrasound propagation velocity, and Durosokop surface strength, so the initial, pre-heat-treatment, and post-heat-treatment results could be compared, not just between the post-heat-treated groups (Fig. 3(a) and (b)).

3 Results

After heat treatment, the monzogranite samples underwent visible and measurable physical changes. In addition to the parameterized factors, the colors and texture of the specimens were also transformed (Fig. 4). A gray-dark gray base color characterizes the monzogranite at 22 °C. In contrast, after heat treatment at 250 °C, small yellowish-brown spots appeared on the surface of the specimens. After the thermal treatment from 22 °C to 500 °C, the whole sample had a pale yellowish-brown base color. The most significant transformation occurred at 750 °C of thermal treatment. The specimens showed a yellowish-reddish-brown color transition, and visible

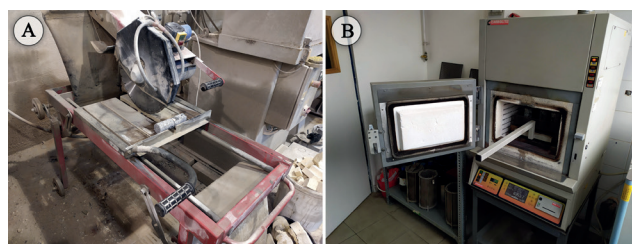


Fig. 2 Instruments for preparation of the monzogranite specimens. a) Cutting of the drill cores, b) Thermal treating in an electric oven

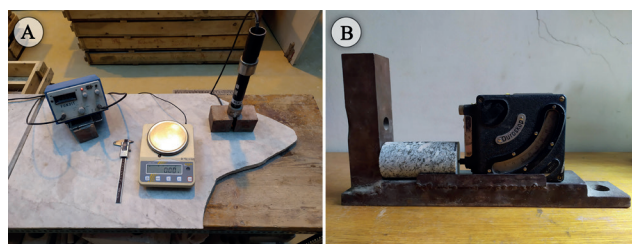


Fig. 3 Instruments for testing the monzogranite specimens a) Bulk density and P-wave velocity measurements, b) Durosokop test

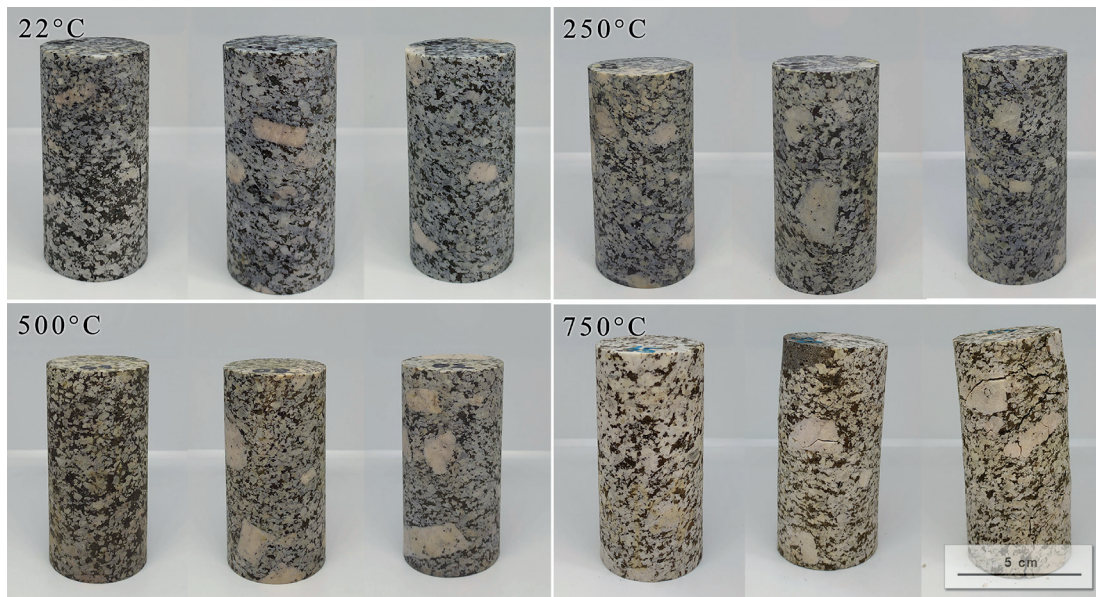


Fig. 4 Thermal-treated cylindrical samples of the porphyritic monzogranite rock formation. Color change is more significant at higher temperatures

macro-cracks appeared on the surface of the samples. Among the primary phases of the rock, the whitening of the gray and light pale pink feldspars appeared, thus separating the light quartz and feldspar from the biotitic phases transformed them into dark red (Fig. 4).

In terms of measured physical parameters, the mean bulk density value at room temperature (22 °C) is 2714.2 kg/m³. After 250 °C heat-treatment, 1.28% decrease-, at 500 °C 0.82% decrease- and at 750 °C heat-treatment, 8.03% decrease in bulk density mean values occurred (Table 1).

The heat-treated bulk density values show that at 500 °C, the bulk density decrease is slighter than the decrease at 250 °C; however, at 750 °C, a significant reduction of bulk density values occurs (Fig. 5).

Table 1 The sample mean and standard deviation results of physical parameters before heat-treatment (22 °C) and after (250 °C, 500 °C, and 750 °C) heat-treatment

Thermal treatment	Bulk density (Std)		P-wave velocity (Std)		Duroskep value (Std)	
	[kg/m ³]		[km/s]		[-]	
	before	after	before	after	before	after
22 °C	2716.0 (23.6)	-	5.29 (0.25)	-	51 (0.4)	-
250 °C	2712.9 (7.5)	2678.1 (34.0)	5.29 (0.15)	4.49 (0.16)	51 (0.5)	49 (0.4)
500 °C	2716.9 (22.9)	2694.7 (27.1)	5.09 (0.36)	3.32 (0.20)	50 (0.8)	47 (0.7)
750 °C	2710.9 (5.7)	2493.3 (106.1)	5.26 (0.16)	0.76 (0.25)	51 (0.8)	19 (5.9)

P-wave velocity shows a uniform decrease after different thermal-treatment processes. The average of 5.22 (0.24) km/s P-wave velocity at room temperature significantly decreases after heat treatment. After 250 °C, 2.49% decrease in velocity-, after 500 °C of treatment, 34.41% and after 750 °C of heat-treatment, 85.69% of P-wave velocity decrease occurred compared to room temperature values (Fig. 6).

The average Duroskep rebound values of the untreated samples are 51, with a 0.8 standard deviation. As the results show, the surface strength slightly decreases to 49 at 250 °C, 47 at 500 °C, and then significantly drops to 19 at 750 °C.

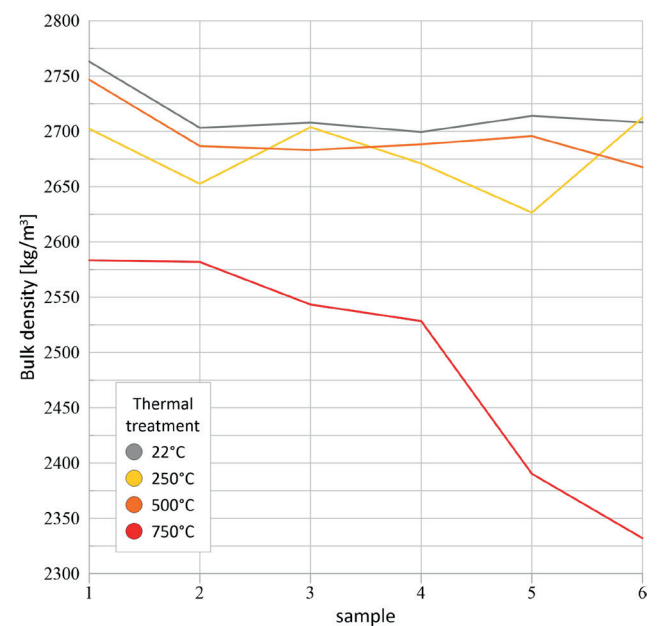


Fig. 5 Line plot of Bulk density values

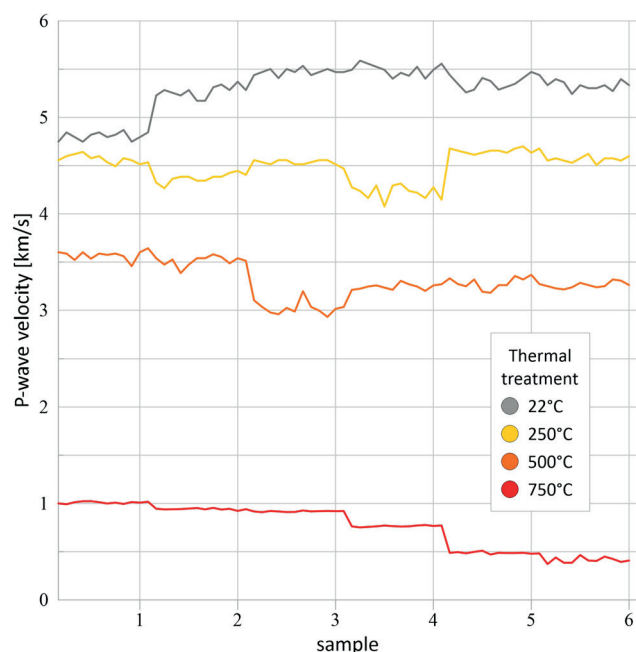


Fig. 6 Line plot of P-wave velocity values

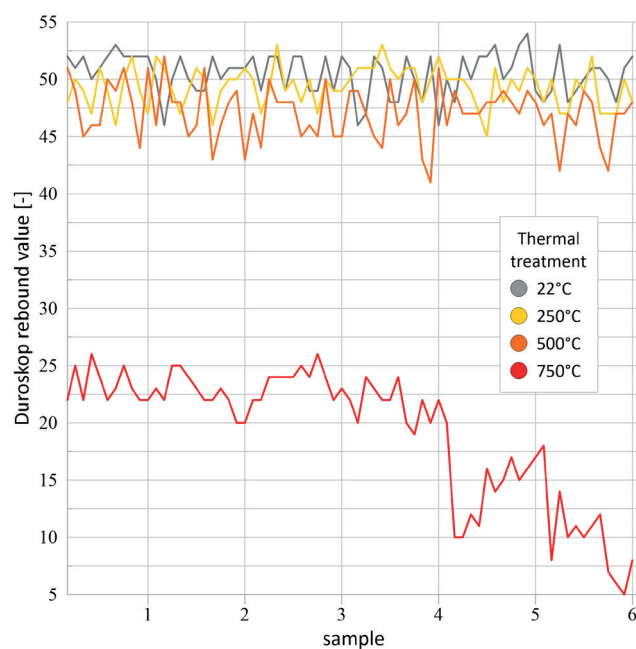


Fig. 7 Line plot of Duroskop values

Duroskop values tend to scatter more than bulk density and P-wave velocity values (Table 1). The measurement method is made on a point-by-point basis, and the rebound value of individual minerals behaves differently at room temperature and after heat treatment (Fig. 7).

It is possible to determine the P-wave modulus of elasticity values (M) for each sample from bulk density and P-wave velocity results [11, 20] (Fig. 8). Results show a linear decrease from standard room temperature to elevated temperatures. P-wave modulus shows a linear correlation with temperature with a high R-value of 98%.

4 Discussion

Non-destructive material testing methods are suitable for detecting physical alterations of the monzogranite samples at elevated temperatures.

Heat-treatment of the monzogranite samples visibly changed the color of the specimens at elevated temperatures. At high temperatures (as 750 °C), not just micro-, but macro-cracks developed due to mineral alterations and the uneven thermal expansion of the individual minerals [11, 21, 22]. These processes changed the mass, volume, bulk density, ultrasonic pulse velocity, and rock surface hardness (Fig. 9).

Heat-treatment processes carried out slight mass decreases and significant volume increase at higher temperatures. Therefore, the parameter is driven mainly by the volume change of the tempering.

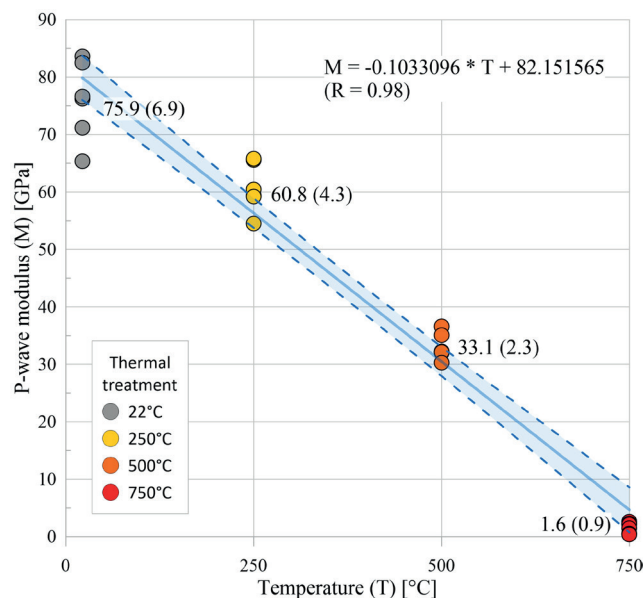


Fig. 8 P-wave modulus (M) of the standard and heat-treated granite samples with the mean values and standard deviations. The results show linear correlation with an R of 98%

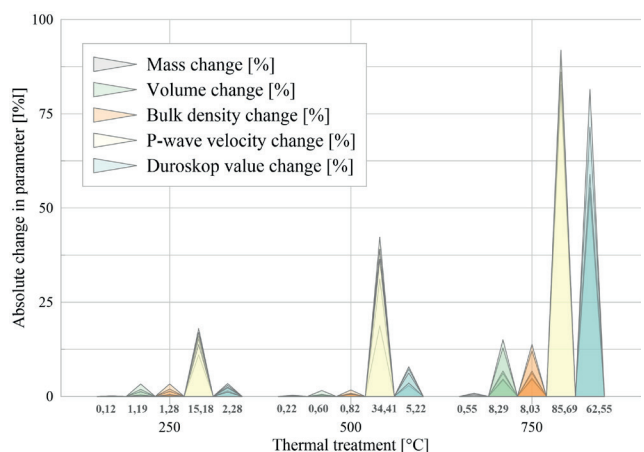


Fig. 9 Absolute data Bar Plot of sample groups, after heat treatment

Bulk density values decrease at 250 °C followed by a minor increase at 500 °C and a significant decrease at 750 °C. Heated samples have lower densities than non-heated ones.

The P-wave velocities clearly show that heat-treatment rapidly and uniformly decreasing ultrasonic pulse values. Former papers suggest [11, 21] that micro-cracks are formed during the heating of granitic rocks. Therefore, slower velocities occur at higher temperatures. P-wave velocity seems to be the best way to indicate the thermal behavior of the monzogranite as all measured results show distinguished and continuously decreasing data between heat-treated groups.

Duroskop results show that surface strength decreases significantly at higher temperatures; however, from 250-500 °C thermal treatment, slight change occurs between groups. The scatter of the Duroskop data suggests that the method is less sensitive than the P-wave velocity values as different crystals tend to have different rebound values.

In terms of P-wave modulus, it can be stated that the specimens gradually lose their elasticity with increasing temperature. The M-modulus is a function of the 2nd power of the P-wave velocity; therefore, it is easy to see that this loss of elasticity is strongly related to the decrease in P-wave propagation velocities, and thus to the formation of micro- and macro-fractures at high temperatures [11, 21, 22]. As for the P-wave velocity results, it can be stated that the M-modulus is one of the best ways indicating thermal-treatment-related physical degradations in the monzogranite.

In the term of combined bulk density, P-wave velocity and Duroskop values show, that the most significant change in both three values tends to decrease significantly between 500 °C and 750 °C. As the ternary class scatters (Fig. 10) indicate, relative percentages of the examined physical parameters form a distinct population at 750 °C from 250 °C and 500 °C. Between 500 °C and 750 °C mineral alterations and related expansions (such as α -quartz to β -quartz inversion at 573 °C) and the thermal expansion difference of main rock-forming minerals cause micro-, and macro cracking inside and on the outer surface of the specimens [11, 21, 22]. This cracking development significantly and permanently increases volume, decreases bulk density-, P-wave velocity- and Duroskop surface strength values. The decrease in ultrasonic pulse velocity at elevated temperatures has been observed at granites [22] and concrete [23].

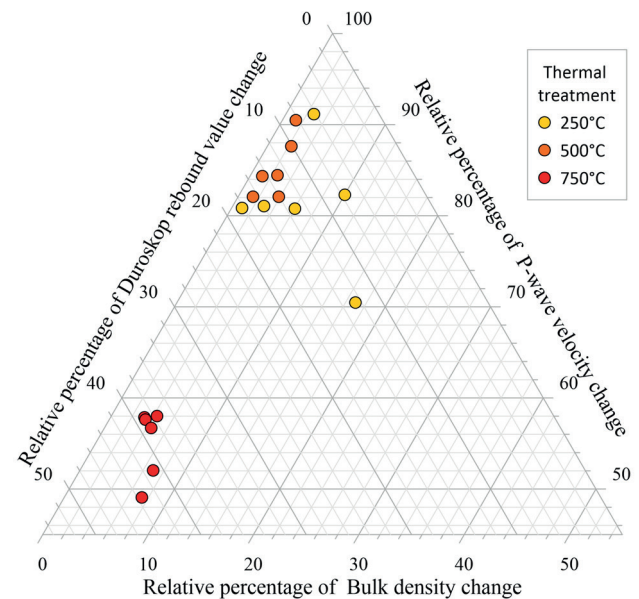


Fig. 10 Ternary class scatter plot of bulk density-, P-wave velocity-, and Duroskop value changes

For further research, the resolution of heat-treatment between 500 °C and 750 °C is inevitable to reveal the exact limit of physical decrease to the host rock.

5 Conclusions

Monzogranite, the host rock of the National Radioactive Waste Storage Facility of Hungary, suffered alteration due to thermal experiments at 250 °C, 500 °C, and 750 °C.

Heat-treatment caused brownish, then reddish color changes in the samples, which is more prominent at higher temperatures. At 750 °C, macro-cracks appeared on the surface of the specimens. Mineral changes and thermal expansion differences of main rock-forming minerals caused a change in the physical properties of the monzogranite samples.

Bulk density values tend to decrease at 250 °C then increase at 500 °C. At 750 °C drastic decrease occurs in values, which is connected to the volume change in the samples.

P-wave velocity and the highly connected P-wave modulus (M) reduction due to high temperature is uniform and significant and seems to be the best way to indicate the thermal behavior of the monzogranite of the examined physical properties. P-wave velocity change is connected to the micro-cracking development of the specimens.

Duroskop values decrease slightly until 500 °C, then significantly at 750 °C. Surface strength changes can be assessed with Duroskop at elevated temperatures, but it

seems very sensitive to smaller-scale surface strength changes, and larger individual crystals can influence the rebound values.

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